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**Non-monochromatic ionization effects induced by ultrashort laser pulses in dielectric crystals**

**Vitaly Gruzdev**  
**MISSOURI SYSTEM, UNIVERSITY OF**

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**Final Report**

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# **Non-Monochromatic Ionization Effects Induced by Ultrashort Laser Pulses in Dielectric Crystals**

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Final Scientific Report

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**Submitted to:** Air Force Office of Scientific Research

ATTN: Dr. Arje Nachman

875 North Randolph Road

Arlington, VA 22205

E-mail: arje.nachman@us.af.mil

**Submitted by:**

University of Missouri

College of Engineering

Department of Mechanical and Aerospace Engineering

Columbia, MO, 65211

**Principal Investigator:** Dr. Vitaly Gruzdev

E-mail: vgruzdev@unm.edu

**Current affiliation:** University of New Mexico

Department of Physics and Astronomy

Albuquerque, NM, 87131

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## ABSTRACT

The objective of this effort is to derive analytic relations for the rates of laser-driven ionization, linear and nonlinear absorption to simulate ultrafast optical response of a dielectric crystal to action of an ultrashort high-intensity laser pulse. Laser-induced ionization is the fundamental process directly associated with absorption of laser-pulse energy via linear (by laser-generated electron-hole plasma) and nonlinear (multiphoton absorption attributed to electron excitation from valence to conduction band) mechanisms. The ionization is one of the key effects affecting ultrafast nonlinear propagation in dielectric solids. The most popular approaches to simulate the ionization either numerically solve some quantum-mechanical equations or solve a rate equation for free-electron density with rates given by monochromatic-approximation analytical relations. Delivering scaling with laser and material parameters (e. g., wavelength, carrier-envelope phase, pulse width, band gap, and effective electron mass) is a challenge for the numerical approaches. The rate-equation models cannot properly treat the ultrashort pulses because significant bandwidth of the pulses does not fit the monochromatic approximation of the models.

This three-year effort has delivered a non-contradictive, non-perturbative analytical model of the ultrafast ionization and nonlinear absorption. The non-perturbative approach considers electron transitions between laser-distorted states under the assumption of low electron-particle collision rate. The distortions are included via the model of laser-driven electron oscillations and result in modification of energy bands involved into the electron excitations. We first derived analytical formulas for the laser-modified energy gaps and energy bands. Then we obtained analytical relations for the rate of photo-ionization (i. e., electron transitions driven by direct action of laser-pulse electric field). The relations were derived for either total photoionization yield by a single-cycle pulse or time-dependent ionization rate by a multi-cycle pulse (at least 3 cycles per pulse). The latter case considered cycle-averaged rates, assumed the photoionization and intra-band excitation of free carriers were decoupled, and was reduced to the Keldysh monochromatic model by assuming zero ratio of cycle duration to pulse width. The relations contain explicit scaling with laser and crystal parameters. The most common model of material response, i. e., a rate equation for the total conduction-band electron density that combined the Keldysh formula for the photoionization rate with the high-collision-rate Drude model of energy absorption by free electrons was revised. The revision considers a rate equation for energy-resolved electron density in the conduction band and the low-collision-rate Vinogradov equation for the rate of energy absorption by oscillating conduction-band electrons. We have derived analytic relations for electron-particle collision rates by use of the cycle-averaged approximation for the laser-perturbed electron states. Obtained results allow full time-dependent non-perturbative description of nonlinear and linear absorption, electron excitation, and transient optical response. Two new physical effects are predicted by the model: generation of bias-free cycle-averaged photocurrent and amplification of weak probe pulses in pump-probe experiments.

Two papers were published in *Physical Review B* and *JOSAB*, three 2-page papers - in the CLEO (Conference on Lasers and Electro-Optics) proceedings, and two 4-page papers - in proceedings of URSI conference. Four papers are under review in *Phys. Rev. B* and *Phys. Rev. Letters*. Three papers are under preparation. 14 oral and poster presentations including 3 invited talks were delivered at international conferences. Obtained results were reported at 4 seminars at University of New Mexico, University of Central Florida - CREOL, University of Rochester - Laboratory for Laser Energetics, and Center for High Technology Materials – University of New Mexico.

## 1. SUMMARY

### 1.1. Overview

This AFOSR funding fully supported the following items within the three year of this research (August 1, 2015 through July 31, 2018):

- salary of a postdoc:

- 10 months of year 1;
- 10 months of year 2;
- 10 months of year 3;

- salary of the PI:

- 2 months of year 1;
- 3 months of year 2;
- 6 months of year 3;

- domestic travels to international conferences:

- 2 trips in year 1 (SPIE Laser Damage symposium – September 2015; High Power Laser Ablation conference – April 2016);
- 2 trips in year 2 (SPIE Laser Damage symposium – September 2016; CLEO – Conference on Lasers and Electro-Optics – May 2017);
- 2 trips in year 3 (SPIE Laser Damage Symposium – September 2017; CLEO – Conference on Lasers and electro-Optics – May 2018);

- international trips:

Domestic (within the US) and international flight of a trip of Dr. Shcheblanov to visit group of Dr. A. Couairon – year 1;

A travel to international conference on fundamentals of Laser Assisted Micro- and Nano-Technologies to deliver an invited talk – year 1;

- purchase of two desk-top computers for simulations – year 1.

Results of the research supported by this grant have been disseminated in two papers published in *Physical Review B* and *JOSA B*, two 4-page conference papers published in the proceedings of URSI-GASS-2017 conference, and three 2-page conference papers published in the proceedings of CLEO – Conference on Lasers and Electro-Optics. Of those 7 published papers, 4 were fully supported, and 3 were partly supported by this award. Of the four submitted manuscripts listed below in section 1.2.3, three were fully and one was partly supported by this award. Of the three papers under preparation listed in section 1.2.4, all were fully supported by this grant.

Results were also disseminated in 3 invited talks, 8 regular oral talks, and 3 poster presentations delivered at CLEO – Conference on Lasers and Electro-Optics, URSI-GASS conference, SPIE Laser Damage Symposium, High Power Laser Ablation conference, and international conference on fundamentals of Laser Assisted Micro- and Nano-Technologies. The conference presentations listed below in section 1.3 were fully supported by this grant. The conference presentations and publications are summarized below in section 1.2 and 1.3.

In addition, the major results of this research project were disseminated at 4 institution- and department-wide seminars delivered at the following institutions:

- College of Optics and Photonics (CREOL), University of Central Florida, Orlando, FL, 7/5/18;
- Department of Physics and Astronomy, University of New Mexico, Albuquerque, NM, 7/23/18

- Center for High Technology Materials (CHTM), University of New Mexico, Albuquerque, NM, 09/28/2018;

- Laboratory of Laser Energetics, University of Rochester, Rochester, NY, 10/26/2018.

Trips to those seminar talks were not supported from this grant and were partly supported by the inviting institutions. However, the presentations essentially contained results obtained under a support from this award. Because of this reason, the support from this award was acknowledged at the end of each seminar talk.

This grant supported establishing domestic research collaboration with the following groups:

- **domestic 1:** Dr. S. Tochitsky, University of California in Los Angeles, Los Angeles, CA, USA (interested in numerical simulations of transient variations of nonlinear optical response by pico- and femto-second laser pulses in several semiconductors);
- **domestic 2:** Dr. A. Sandhu, University of Arizona, Tucson, AZ, USA (interested in numerical simulations of transient optical response to support pump-probe experiments);
- **domestic 3:** Dr. Igal Brener, Sandia National Lab, Albuquerque, NM (interested in advanced simulations of ultrafast nonlinear response of nanostructures);
- **domestic 4:** Dr. Wolfgang Rudolph, University of New Mexico, Albuquerque, NM (interested in experimental verification of theoretically predicted bias-free photocurrent);
- **domestic 5:** Dr. Tsing-Hua Her, University of North Carolina at Charlotte, Charlotte, NC (interested in simulation of ultrafast nonlinear absorption by polymers).

This project synergized collaborations with the group of Dr. Enam Chowdhury from the Ohio State University (OSU) that resulted in ongoing collaborative project supported by another AFORS grant (January 2016 – December 2019).

International research contacts were established with Dr. Arnaud Couairon (Ecole Polytechnique, France). In 2017, I established very promising contacts with Dr. Martin Schultze, Dr. Vladislav Yakovlev, and Dr. Vladimir Pervak from Max Plank Institute for Quantum Optics, Munich, Germany. They were extremely interested in collaboration and invited to visit their institution.

Research activity supported by this grant is briefly described below in several sections structured according to the research tasks of the original project narrative. There is no change in the project objectives. For each task, planned milestones are considered for evaluation of research efforts. Together with successful performance, challenges are discussed.

## **1.2.Publications**

### **1.2.1. Peer-reviewed papers published in journals:**

1. O. Sergaeva, V. Gruzdev, D. Austin, E. Chowdhury, “Ultrafast excitation of conduction-band electrons by high-intensity ultrashort laser pulses in band-gap solids: Vinogradov equation vs Drude model”, *J. Opt. Soc. Am. B*, v. **35** (11), pp.2895-2905 (2018).
2. V. Gruzdev and O. Sergaeva, “Ultrafast modification of band structure of wide-band-gap solids by ultrashort pulses of laser-driven electron oscillations”, *Phys. Rev. B*, v. **98** (11), 115202 (2018).

### 1.2.2. Published peer-reviewed conference papers:

1. V. Gruzdev and O. Sergaeva, "Influence of crystal structure on the ultrafast ionization of cubic wide-band-gap crystals by ultrashort laser pulses," in Conference on Lasers and Electro-Optics, OSA Technical Digest (online) (Optical Society of America, 2018), paper SM3O.4 (May 2018).

URL: [https://www.osapublishing.org/abstract.cfm?uri=CLEO\\_SI-2018-SM3O.4](https://www.osapublishing.org/abstract.cfm?uri=CLEO_SI-2018-SM3O.4)

2. V. Gruzdev, O. Sergaeva, "Ultrafast mechanism of energy-band modification of wide-band-gap crystals by pondermotive potential of Gaussian ultrashort laser pulse," *2017 XXXIInd General Assembly and Scientific Symposium of the International Union of Radio Science (URSI GASS)*, Montreal, QC, 2017, pp. 1-4, doi: 10.23919/URSIGASS.2017.8105005; URL: <http://ieeexplore.ieee.org/stamp/stamp.jsp?tp=&arnumber=8105005&isnumber=8104490>

[http://www.ursi.org/proceedings/procGA17/papers/Paper\\_D7-1\(2548\).pdf](http://www.ursi.org/proceedings/procGA17/papers/Paper_D7-1(2548).pdf)

3. V. Gruzdev, D. Austin, O. Sergaeva, E. Chowdhury, "Simulations of ultrafast laser-induced excitation and heating of electron sub-system of semiconductors with the Vinogradov equation and multi-band Keldysh formula," *2017 XXXIInd General Assembly and Scientific Symposium of the International Union of Radio Science (URSI GASS)*, Montreal, QC, 2017, pp. 1-4; doi: 10.23919/URSIGASS.2017.8105007; URL: <http://ieeexplore.ieee.org/stamp/stamp.jsp?tp=&arnumber=8105007&isnumber=8104490>

[http://www.ursi.org/proceedings/procGA17/papers/Paper\\_D7-2\(2547\).pdf](http://www.ursi.org/proceedings/procGA17/papers/Paper_D7-2(2547).pdf)

4. V. Gruzdev, D. R. Austin, O. N. Sergaeva, E. Chowdhury, "Beyond the Drude Approach: a Keldysh-Vinogradov Model of Dynamics of Ultrafast Laser-Induced Electron Excitation", in *Conference on Lasers and Electro-Optics*, OSA Technical Digest (online) (Optical Society of America, 2017), paper STh4J.6, May 2017.

URL: [https://www.osapublishing.org/abstract.cfm?uri=CLEO\\_SI-2017-STh4J.6](https://www.osapublishing.org/abstract.cfm?uri=CLEO_SI-2017-STh4J.6)

5. O. N. Sergaeva, V. Gruzdev, "Modification of Energy Bands of a Dielectric Crystal by Pondermotive Potential of Gaussian Ultrashort Laser Pulse", in *Conference on Lasers and Electro-Optics*, OSA Technical Digest (online) (Optical Society of America, 2017), paper JTh2A.28, May 2017.

URL: [https://www.osapublishing.org/abstract.cfm?uri=CLEO\\_SI-2017-JTh2A.28](https://www.osapublishing.org/abstract.cfm?uri=CLEO_SI-2017-JTh2A.28)

### 1.2.3. Submitted manuscripts:

1. V. Gruzdev, O. Sergaeva, W. Rudolph, "Bias-free ultrashort pulses of photocurrent driven by few-cycle laser pulses in non-metal crystals: theoretical analysis", *Phys. Rev. Lett.* (submitted, under review).
2. V. Gruzdev, O. Sergaeva, D. Austin, E. Chowdhury, "Multi-band non-perturbative Keldysh-Vinogradov model of ultrafast laser-induced excitation of electron-hole plasma in wide-band-gap crystals", *Phys. Rev. B* (submitted; under review).
3. V. Gruzdev, O. Sergaeva, "Ultrafast mechanisms of band-structure modification in wide-band-gap crystals", *Opt. Eng.* (submitted, under review).
4. V. Gruzdev, O. Sergaeva, "Ultrafast modification of cosine energy bands of wide-band-gap

crystals by ultrashort laser pulses”, *Phys. Rev. B* (submitted, under review).

#### 1.2.4. Manuscripts under preparation:

1. V. Gruzdev, O. Sergaeva, “Ultrafast modification of cosine energy bands of wide-band-gap solids by ultrashort pulses of laser-driven electron oscillations” (for submission to *Phys. Rev. B*).
2. V. Gruzdev, O. Sergaeva, “Time-dependent non-perturbative Keldysh-type model of the photoionization of dielectric crystals by few-cycle laser pulses: parabolic-band approximation” (for submission to *Phys. Rev. B*).
3. V. Gruzdev, O. Sergaeva, “Non-perturbative collision integrals for electron-particle collisions driven by high-intensity ultrashort laser pulses” (for submission to *Phys. Rev. B*).

### 1.3. Conference Presentations

#### A. Invited talks:

- 1) V. Gruzdev, O. Sergaeva, “Band-structure modification of wide-band-gap crystals by laser-driven Bloch oscillations of electrons: monochromatic vs non-monochromatic effects”, The International High-Power Laser Ablation Conference VIII (HPLA-20), Santa Fe, NM, USA, 26 – 29 March 2018;
- 2) V. Gruzdev, O. Sergaeva, “Ultrafast mechanism of energy-band modification of wide-band-gap crystals by pondermotive potential of Gaussian ultrashort laser pulse”, 32<sup>nd</sup> International Union of Radio Science General Assembly and Scientific Symposium (URSI GASS-2017), Montreal, Canada, 19-26 August 2017;
- 3) V. Gruzdev, N. Shcheblanov, “Perspectives of theoretical models of laser photoionization of non-metal crystals: from the Keldysh approximation to time-dependent theory”, International Symposium “Fundamentals of Laser Assisted Micro- and Nanotechnologies” (FLAMN-16), St. Petersburg, Pushkin, Russia, June 27 - July 1, 2016.

#### B. Regular oral presentations:

- 1) V. Gruzdev, “The photo-ionization and band structure of solids: non-trivial interplay”, SPIE Laser Damage Symposium, Boulder, CO, USA, 27-30 September 2015;
- 2) V. Gruzdev, “Laser-induced photoionization: atoms vs solids”, High Power Laser Ablation and Directed Energy conference, Santa Fe, NM, USA, 4-7 April 2016;
- 3) V. Gruzdev, D. R. Austin, E. A. Chowdhury, “Influence of multiple energy bands on the photoionization of non-metal crystals”, SPIE Laser Damage Symposium, Boulder, CO, USA, 27-30 September 2016;
- 4) V. Gruzdev, D. R. Austin, O. N. Sergaeva, E. Chowdhury, “Beyond the Drude Approach: a Keldysh-Vinogradov Model of Dynamics of Ultrafast Laser-Induced Electron Excitation”, CLEO 2017: Science and Innovations, San Jose, CA, USA, 14-19 May 2017;
- 5) V. Gruzdev, D. Austin, O. Sergaeva, E. Chowdhury, “Simulations of ultrafast laser-induced excitation and heating of electron sub-system of semiconductors with the Vinogradov equation

and multi-band Keldysh formula”, 32<sup>nd</sup> International Union of Radio Science General Assembly and Scientific Symposium (URSI GASS-2017), Montreal, QC, Canada, 19-26 August 2017;

6) Vitaly E. Gruzdev, Olga Sergaeva, “Conversion of direct-gap energy bands of a wide-band-gap crystal into indirect-gap transient bands by ponderomotive potential of Bloch oscillations driven by ultrashort laser pulse”, SPIE Laser Damage Symposium, Boulder, CO, USA, 24-27 September 2017;

7) Drake R. Austin, Kyle R. P. Kafka, Yu H. Lai, Zhou Wang, Kaikai Zhang, Hui Li, Cosmin I. Blaga, Allen Y. Yi, Louis F. DiMauro, Vitaly E. Gruzdev, Enam A. Chowdhury, “Wavelength dependence of the mid-IR ablation threshold of ZnSe”, SPIE Laser Damage Symposium, Boulder, CO, USA, 24-27 September 2017;

8) V. Gruzdev and O. Sergaeva, "Influence of crystal structure on the ultrafast ionization of cubic wide-band-gap crystals by ultrashort laser pulses", Conference on Lasers and Electro-Optics (CLEO), San Jose, CA, USA, 13-18 May 2018.

### C. Poster presentations:

1) O. N. Sergaeva, V. Gruzdev, “Modification of Energy Bands of a Dielectric Crystal by Ponderomotive Potential of Gaussian Ultrashort Laser Pulse”, CLEO 2017: Science and Innovations, San Jose, CA, May 14-19, 2017;

2) Vitaly E. Gruzdev, Olga Sergaeva, Drake Austin, Enam A. Chowdhury, “Beyond the Drude model: the Keldysh-Vinogradov model of ultrafast generation and heating of electron-hole plasma”, SPIE Laser Damage Symposium aka XLIX Annual Symposium on Optical Materials For High Power Lasers, Boulder, CO, USA, 24-27 September 2017;

3) O. Sergaeva, V. Gruzdev, D. R. Austin, E. Chowdhury, “From Keldysh-Drude model to Keldysh-Vinogradov model: Fixing of some fundamental issues”, The International High-Power Laser Ablation conference (HPLA-20), Santa Fe, NM, USA, 26 – 29 March 2018.

## **1.4. Research contacts**

*1.4.1. Domestic research contacts.* The support by this award have resulted in establishing domestic research collaboration with research teams from six US universities.

*1) Group of Dr. S. Tochitsky, University of California in Los Angeles, Los Angeles, CA, USA.*  
[http://pbpl.physics.ucla.edu/Research/Experiments/Advanced\\_Accelerators/IFEL/People/](http://pbpl.physics.ucla.edu/Research/Experiments/Advanced_Accelerators/IFEL/People/)  
<http://www.seas.ucla.edu/plasma/people.html>

This team experimentally studies nonlinear optical response of semiconductors to pico- and femto-second laser pulses of mid-infrared wavelength. They are supported by AFOSR via the program of Riq Parra. Their recent experimental data are not explained by the usual models of ultrafast laser-solid interactions. They are highly interested in simulation of their experiments with the novel approaches I reported at CLEO-2017 conference. Our contacts are successfully progressing, and I have received some experimental data for test simulations.

*2) Group of Dr. A. Sandhu, University of Arizona, Tucson, AZ, USA.*

<http://www.optics.arizona.edu/research/faculty/profile/arvinder-sandhu>  
<http://www.physics.arizona.edu/~sandhu/>

The contact with this group was established right after my talk at CLEO-2017. This team made pump-probe experiments on several samples of topological insulators and observed unusual dynamics of reflectivity signal. The traditional theoretical models do not explain those results. I have received some experimental data from Dr. Sandhu for a test simulation.

*3) Group of Dr. Igal Brener, Sandia National Lab, Albuquerque, NM, USA.*  
<https://www.lanl.gov/expertise/profiles/view/igal-brener>

This team works in the field of ultrafast switching of nanostructured surfaces (metaoptics surfaces). Materials of the nanostructures are typical semiconductors, and their excitation by ultra-short laser pulses is well described by the model we have developed under support of this award. I have received experimental data for a test simulation. Contacts are in progress.

*4) Dr. Wolfgang Rudolph, University of New Mexico, Albuquerque, NM, USA.*  
<http://www.phys.unm.edu/opsci/wr/>

This team expressed interest in performing proof-of-concept experiments to detect the ultrashort pulses of bias-free photocurrent we recently predicted by our analytical model. Dr. Rudolph has a solid expertise in experimental studies of traditional photocurrents generated by femtosecond laser pulses. The team has access to femtosecond lasers that meet the theoretical estimations for proof-of-concept experiments. We are working on preparation of a white paper on this research.

*5) Dr. Tsing-Hua Her, University of North Carolina at Charlotte, Charlotte, NC, USA.*  
<https://physics.uncc.edu/people/tsing-hua-her>  
<https://pages.uncc.edu/tsing-hua-her/>

They are interested in simulation of nonlinear absorption of ultrashort laser pulses by polymers to support their recent experiments on two-pulse ultrafast laser ablation. I have received some of their data for test simulations. Our contacts are progressing towards a joint paper.

*6) I especially emphasize research collaboration with Dr. Enam Chowhury (The Ohio State University, Columbus, OH) that was substantially initiated due to this award. It has resulted in joint research supported by AFOSR via the program of Riq Parra. I provide a theoretical support for the experiments done by the team of Dr. Chowhury. The model developed for Tasks 2 and 3 under this award turns out to deliver a very good approximation for their experimental data obtained with mid-infrared femtosecond laser pulses. The collaboration with that team is very favorable for expansion of applications of the models proposed and developed under support from this research award.*

*1.4.2. International research contacts* established under support of this award are as follows:

A) The research contacts with Dr. Arnaud Couairon (Ecole Polytechnique, France) began in late 2016. As a part of this effort, a visit of Dr. Shcheblanov to Dr. Couairon was partly supported by this award in early 2016. However, our contacts degraded in 2016-2017. [REDACTED]

[REDACTED]  
[REDACTED]  
[REDACTED] In the future the contacts can be re-activated.

B) In September 2017, I established research contacts with Dr. M. Schultze (leader of semiconductor group), Dr. V. Yakovlev (leader of theoretical group), and Dr. V. Pervak (leader of ultrafast optics group) from Max Plank Institute of Quantum Optics in Munich, Germany. They attended my oral presentation on laser-induced modification of energy bands by ultrashort laser pulses. I received an invitation to visit Max Plank Institute of Quantum Optics in spring 2018 to deliver a seminar talk and discuss research collaborations. They also kindly agreed to support my trip to their institution. However, my unemployment by University of Missouri in April 2018 completely changed those plans. My visit was re-scheduled for late spring 2019. Contacts with those group leaders are in progress.

## PERFORMANCE BY TASK AND BY MILESTONE

### **Task 1. Analytical derivation of the time-dependent photo-ionization rate and conduction-band electron density.**

#### YEAR 1

- a) Formulation of the general problem and derivation of general relations for the probability amplitude and transition matrix element;
- b) Derivation and analysis of laser-modified energy bands for several specific models of pulse envelopes and energy-momentum relations;
- c) **Derivation of the photo-ionization yield and instant rate for the multi-frequency model.**

#### YEAR 2:

- d) General evaluation of the time-dependent conduction-electron density for the first asymptotic limit – pulse duration as short as 1 period of field oscillations;
- e) Evaluation of the first asymptotic limit for particular energy-momentum relations (cosine and more complicated realistic energy bands);

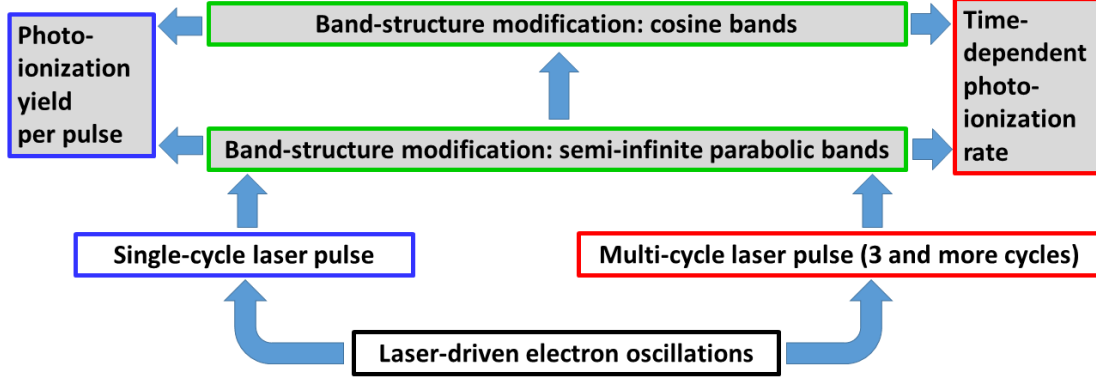
#### YEAR 3:

- f) General evaluation of the time-dependent conduction-electron density and time-dependent instant ionization rate for the second asymptotic limit – pulse duration is much longer than 1 period of field oscillations;
- g) Evaluation of the second asymptotic limit for particular energy-momentum relations (cosine and more complicated realistic energy bands).

=====

This task was almost completely done and all milestones were reached except those related to multi-frequency radiation. In the project narrative, I proposed to make a transition from single-frequency (i. e., the monochromatic approximation) to multi-frequency case (i. e., a coherent superposition of several monochromatic pulses at different frequencies) and then – to a broad-bandwidth pulse. The transition was assumed to employ Fourier transformation and analysis of the entire problem in frequency domain. On course of calculations, it turned out that a general case of an ultrashort laser pulse with a substantial bandwidth can be analytically treated in time domain in an easier way without making the complicated transition via the multi-frequency case. Another reason to skip the milestone C was the poor performance of a postdoc supported by this award in the first year of the project. Since he delivered no outputs, I did have to rearrange the research plan for years 2 and 3 in such a way as to minimize the impact of the first year on the overall performance and successfully reach other milestones. Due to those reasons, the multi-frequency case was not considered. Two cases were considered in analytical calculations: a single-cycle (case I) and a multi-cycle (3 or more cycles; case II) pulse. The final deliverables from this effort that go to the other tasks of this project include (Fig. 1):

- I) analytical formulas for time-dependent laser-modified energy bands for both parabolic and cosine models of energy bands for a single-cycle laser pulse;
- II) analytical formulas for time-dependent laser-modified energy bands for both parabolic and cosine models of energy bands for a multi-cycle laser pulse;
- III) analytical formulas for total photoionization yield per pulse for both parabolic and cosine models of energy bands for a single-cycle laser pulse;
- IV) analytical formulas for time-dependent photoionization rate for both parabolic and cosine models of energy bands for a multi-cycle laser pulse.



**Fig. 1.** A diagram of research approach for Task 1. The blocks with grey filling show successfully reached milestones and final results.

Deliverables I are under preparation for submission to *Phys. Rev. B*. Deliverables II either have been published (the model of parabolic band) or have been submitted (the model of cosine band). Deliverables III and IV are under preparation for submission to peer-reviewed journals. The current stage of preparation is generation of figures; checking of equations; revision of the first draft. Planned date for submission of the manuscripts with deliverables I, III, and IV is spring 2019. In December 2019, those deliverables will be submitted to the Conference on Lasers and Electro-Optics (CLEO-2020). 2-page conference papers will be published in CLEO proceedings.

### **Overview of technical approaches**

The problem under consideration was treated in a non-perturbative way by considering 3D time-dependent Schrodinger equation in length gauge:

$$i\hbar \frac{\partial \psi(\vec{r}, t)}{\partial t} = H_0(\vec{r})\psi(\vec{r}, t) - q\vec{E}(t)\vec{r} \quad (1)$$

where  $E(t)$  is instant electric field of a linearly polarized laser pulse. A zero-order approximation for solutions to this equation are the Bloch functions:

$$\psi(\vec{r}, t) = u_{CB}(\vec{p}[t], \vec{r}) \exp\left(\frac{i}{\hbar} \vec{p}[t] \cdot \vec{r}\right) \exp\left(-\frac{i}{\hbar} \int_{-\infty}^t \varepsilon_{CB}(\vec{p}[\tau]) d\tau\right), \quad (2)$$

with time-dependent crystal momentum:

$$\vec{p}(t) = \vec{p}_0 - q \int_{-\infty}^t \vec{E}(\tau) d\tau \quad (3)$$

(see details in V. Gruzdev, O. Sergaeva, PRB, v. 98 (11), 115202, 2018).

The laser-modified functions of Eq. (2) are employed to evaluate probability of laser-driven electron transitions from valence to conduction band and modification of electron energy spectrum. Evaluation of the laser-induced modifications of energy spectrum (i. e., energy bands or band structure) in 3D energy-momentum space is considered as a separate fundamental problem since it is involved into Task 2. The modified energy bands are referred to as effective bands and are characterized by effective (i. e., laser modified) energy gaps between them. Evaluation of the effective band gaps is one of the primary sub-tasks of this effort.

#### *1) Single-cycle laser pulse*

Evaluation of transition probability from a laser-modified valence band to a laser-modified

conduction band is done with functions of Eq. (2) in 3D space for two participating energy bands. Since introduction of a transition rate is not reasonable in this case, total transition yield (and, correspondingly, conduction-band population) per entire laser pulse is evaluated. Involved integration can be done by the methods similar to those from atomic theory of ultrafast photoionization (e. g., see S. V. Popruzhenko, *J. Phys. B: At. Mol. Opt. Phys.* **47**(20), 204001 (2014)). In our calculations, we employed time-dependent saddle-point method with imaginary time (and momentum). In this case, the saddle points are time-dependent and move around the points evaluated from the monochromatic approximation. Final results contain explicit dependence of the photoionization yield on laser (peak intensity; carrier-envelope phase; central wavelength; pulse width) and material (band gap; effective electron and hole masses) parameters.

## II) Multi-cycle laser pulse

Electric field is represented by a product of a slow envelope  $f$  and oscillation at carrier frequency:

$$\vec{E}(t) = \vec{F}_0 f\left(\frac{t}{\tau_p}\right) \cos(\omega_0 t + \phi_0), \quad (4)$$

where  $\tau_p$  is some characteristic duration of the laser pulse, and  $\phi_0$  is carrier-envelope phase. The electric field is assumed to be evaluated by proper accounting for all relevant many-body effects in the dielectric solids. Electron excitation is characterized by time-dependent rate of electron promotion over a laser-modified band gap between two specific energy bands in 3D momentum space. The modified band gap and the transition rate are evaluated in time domain by averaging over duration of a single cycle at carrier frequency. All relevant integrals are asymptotically evaluated using the following parameter:

$$\alpha = \omega_0 \tau_p = 2\pi \frac{\tau_p}{T_0} \gg 1, \quad (5)$$

where  $T_0$  is duration of a single cycle at carrier frequency of laser field  $T_0 = 2\pi/\omega_0$ . Therefore, the derived analytical relations describe slow variations of the laser-modified energy bands and photoionization rate that follow time variations of the pulse envelope. For example, the laser-modified parabolic conduction band reads as follows (see details and explicit expressions for the functions  $F1, F2, F3, G1, G2, G3$  in V. Gruzdev, O. Sergaeva, PRB, v. 98 (11), 115202, 2018):

$$\varepsilon_{CB}^{eff}(\vec{p}_0, s) = \Delta \left[ 1 + \frac{p_0^2}{2m_{CB}\Delta} + \frac{f(s)^2}{4\gamma_{CB}^2} + \frac{f'(s)}{\alpha\gamma_{CB}^2} F1 + \frac{1}{\alpha^2\gamma_{CB}^2} F2 + \frac{1}{\alpha^3\gamma_{CB}^2} F3 \right], \quad (6)$$

where the zero-order terms obtained by assuming  $1/\alpha = 0$  correspond to the monochromatic approximation. Here and below,  $\gamma_{CB}$ ,  $\gamma_{VB}$ , and  $\gamma_0$  are the Keldysh adiabatic parameters for the conduction band, valence band, and inter-band transitions correspondingly:

$$\gamma_i = \frac{\omega_0 \sqrt{m_i \Delta}}{q E_0}; \quad i = CB, VB, 0, \quad \frac{1}{m_0} = \frac{1}{m_{CB}} + \frac{1}{m_{VB}}. \quad (7)$$

A similar expression is obtained for the laser-modified parabolic valence band:

$$\varepsilon_{VB}^{eff}(\vec{p}_0, s) = -\Delta \left[ \frac{p_0^2}{2m_{VB}\Delta} + \frac{f(s)^2}{4\gamma_{VB}^2} + \frac{f'(s)}{\alpha\gamma_{VB}^2} G1 + \frac{1}{\alpha^2\gamma_{VB}^2} G2 + \frac{1}{\alpha^3\gamma_{VB}^2} G3 \right] \quad (8)$$

Similar asymptotic expansions were obtained for cosine energy bands. The rate of the photoionization is represented by some asymptotic series of the following type:

$$W_{PI}(t) = W_{Keldysh} + \frac{1}{\alpha\gamma^2} W_1 + \frac{1}{\alpha^2\gamma^2} W_2 \quad (9)$$

where each term contains exponential dependence on laser-induced effective band gap:

$$W_{PI} \propto \exp(-\Delta_{Deff}[t]) \quad (10)$$

### **Major results**

A completely analytical theory of laser-induced modification of parabolic and cosine energy bands delivers explicit scaling of all major effects with 6 laser and material parameters (central wavelength; peak intensity; pulse width; carrier-envelope phase; band gap; effective masses). As an example, please see V. Gruzdev, O. Sergaeva, PRB, v. 98 (11), 115202 (2018). This result substantially changes the landscape of theoretical research in the field of high-intensity ultrafast effects induced by few-cycle (from 3 to 30 cycles) laser pulses. For example, contributions of the non-monochromatic terms of the asymptotic series of Eqs. (6), (8), and (9) depend on a product of parameter  $\alpha$  and squared Keldysh parameter. That is, the higher intensity, the stronger the non-monochromatic effects. This non-trivial fact suggests that the non-monochromatic effects can be significant for high-intensity pulses even if they contain a few tens of cycles. Moreover, scaling with laser parameters suggests that the non-monochromatic effects become stronger with increase of laser wavelength and reduction of band gap. The most surprising is the influence of carrier-envelope phase: at values  $\pm\pi/2$ , it suppresses the non-monochromatic effects (Fig. 2).

Furthermore, the efforts of Task 1 have delivered **novel physical effects**.

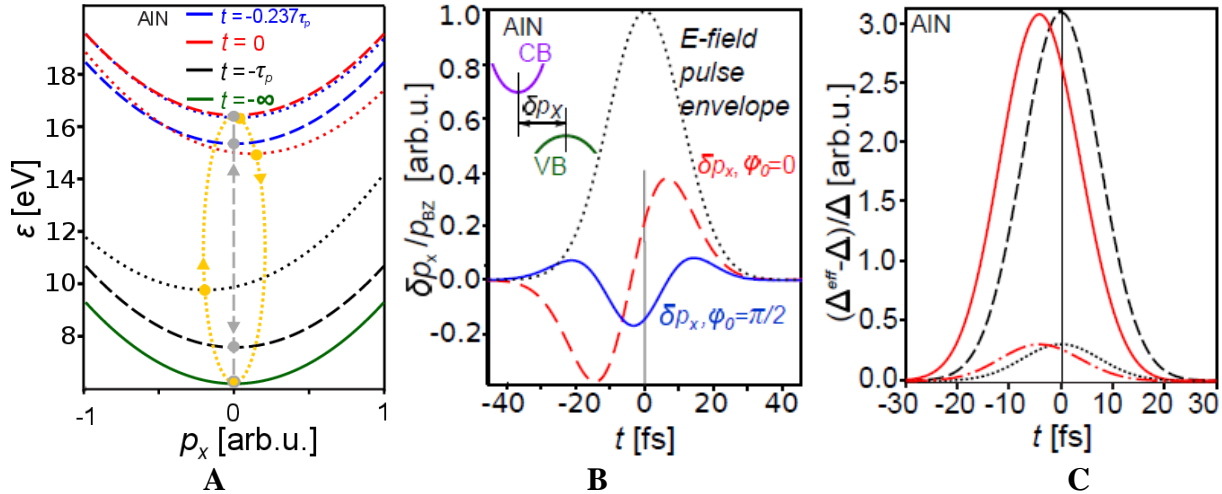
A) It is predicted a shifting of laser-modified energy bands in momentum space that transforms original direct-gap energy bands into indirect-gap energy bands (Fig. 2). Therefore, a direct-gap crystal responds to the ultrafast action of few-cycle pulses as indirect-gap crystal characterized by two time-dependent effective band gaps: direct and indirect. This modification of the energy bands substantially affects both inter-band and intra-band laser-driven electron dynamics.

B) Simulations suggest that the photoionization is substantially suppressed at the leading part of a laser pulse and is enhanced at the tail part of the pulse. This effect results from the exponential dependence of the photoionization rate on effective direct band gap (see Eq. (10)) and the shift of the peak value of effective direct band gap towards the leading part of a pulse (fig. 2). Therefore, the symmetric with respect to pulse peak time evolution of  $N$ -photon absorption characteristic of the monochromatic models is violated: absorption of at least  $N+1$  photons is required at the leading edge of the pulse to bridge the pulse-generated effective band gap, and absorption of  $N-1$  (or even fewer) photons is required at the tail of the pulse. This qualitative analysis is perfectly confirmed by simulations with the non-monochromatic photoionization rate of Eq. (9) and completely changes the current understanding of the dynamics of ultrafast nonlinear absorption.

C) The parabolic bands experience shifting in energy-momentum space without deformation (fig.

2), i. e., an initial value of effective mass does not change. The central part of a laser-distorted cosine band experiences a shift in momentum space coupled to deformation of the band: one side of the band stretches, and the opposite side shrinks. The deformation results in time variations of effective electron (and hole) mass that affects the laser-driven electron dynamics and responses.

D) ***An unexpected and non-trivial side result*** is a prediction of a bias-free photocurrent induced by the specific laser-driven intra-band polarization (V. Gruzdev, O. Sergaeva, PRB, v. 98 (11), 115202, 2018). Compared to the previous publications on a similar effect (A. Schiffrin, T. Paasch-Colberg, N. Karpowicz, et al, *Nature*, **493**, 70 (2013)), our model considers a different photocurrent mechanism and delivers a complete scaling with laser and material parameters.



**Fig. 2.** (A) Original conduction band (green solid) and transient bands (black, blue, red) evaluated for a Gaussian pulse envelope; peak intensity 30 TW/cm<sup>2</sup>; carrier wavelength 2400 nm; pulse half-width at the level 1/e of maximum intensity is 15 fs; zero CEP) at several instants of time for AlN crystals under the monochromatic (dashed lines) and pulse-driven (dotted lines) approximations. Orange dots on the closed dotted curve and gray dots on the vertical gray dashed line depict positions of the bottom of the transient conduction bands on their laser-driven trajectories for the pulse-driven and monochromatic approximations correspondingly. (B) Normalized envelope of E-field pulse (black dotted) and time variations of the total mutual  $p$ -shift of the energy bands  $\delta p_x$  normalized to the half-width of the first Brillouin zone  $p_{BZ}$  of AlN crystal  $0^\circ$  (red dashed) and  $90^\circ$  (blue solid) CEP. The vertical solid lines mark position of the pulse peak on the time axis for eye convenience. (C) Normalized difference between the effective direct band gap and the original band gap for the monochromatic model (black curves) and the pulse-driven model (red curves) plotted vs time at peak laser intensity 10 TW/cm<sup>2</sup> (dashed curves) and 30 TW/cm<sup>2</sup> (solid curves) in AlN. The vertical solid lines mark position of the pulse peak on the time axis for eye convenience. All data are from V. Gruzdev, O. Sergaeva, PRB, v. 98 (11), 115202, 2018.

**Task 2. Analytical derivation of the kinetic coefficients for non-parabolic energy-momentum relations and non-monochromatic light.**

YEAR 1:

- a) Evaluation of electron-electron collision rate for multiple cosine conduction bands in general case;
- b) Evaluation of electron-phonon (optical phonons) collision rate for multiple cosine conduction bands in general case;
- c) Evaluation of electron-defect collision rates for multiple cosine conduction bands in general case;

YEAR 2:

- d) Reduction of obtained non-parabolic results for the case of monochromatic radiation;
- e) **Derivation of the collision rates for parabolic energy-momentum relation and the multi-frequency approximation;**
- f) **Reduction of parabolic-band results for the first asymptotic limit (pulse duration as short as 1 period of field oscillations) and comparison with the monochromatic approximation;**
- g) Reduction of parabolic-band results for the second asymptotic limit (pulse duration is much longer than 1 period of field oscillations) and comparison with the monochromatic approximation;

YEAR 3:

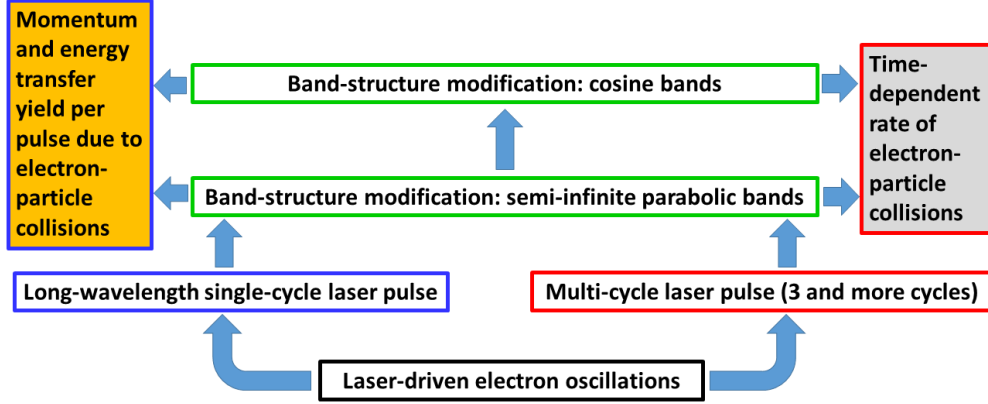
- h) **Reduction of obtained non-parabolic-band collision rates for the first asymptotic limit (pulse duration as short as 1 period of field oscillations);**
- i) Reduction of the general non-parabolic-band results for the second asymptotic limit (pulse duration is much longer than 1 period of field oscillations).

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Of 9 milestones of this Task, six have been successfully reached. The three milestones (marked by red above) are attributed to the collisions driven by a single-cycle laser pulse at wavelength long enough to accommodate multiple collisions. This means a duration of a single cycle must be significantly larger than collision time, i. e., at least 40-50 fs (the shortest time of momentum dephasing in laser-excited solids is about 18-20 fs; see P. C. Becker, et al, *Phys. Rev. Lett.* **61**, 1647 (1988); also B. B. Hu, E. A. de Souza, et al, *Phys. Rev. Lett.* **74** (9), 1689-1692 (1995)). Corresponding wavelength is longer than 15  $\mu\text{m}$  and falls into far-infrared or THz ranges of spectrum that are of minor interest for the proposed research. For shorter wavelengths, multiple electron collisions within a single cycle are not realistic for the high-intensity ultrafast laser-dielectric interactions.

Those facts were taken into account when I was developing a plan to mitigate the impact of the poor performance of the postdoc employed in the first year. Correspondingly, milestone E (multi-frequency case) was skipped together with milestones of the other Tasks focused on the multi-frequency case. Milestone H was skipped by eliminating consideration of the far-infrared and THz parts of the spectrum. A major effort was focused on milestones A-D, G, and I to treat the electron-particle collisions under the monochromatic and multi-cycle-pulsed approximations for parabolic and cosine energy-momentum relations (Fig. 3). Accomplishment of those milestones were successful. The deliverables from this Task for modeling of Task 3 included:

- I) analytical formulas for the rate of electron-electron, electron-optical-phonon, and electron-defect collisions for the approximation of multi-cycle laser pulses and parabolic band;
- II) analytical formulas for the rate of electron-electron, electron-optical-phonon, and electron-defect collisions for the approximation of multi-cycle laser pulses and cosine band.



**Fig. 3.** A diagram of research approach for Task 2. The blocks with grey filling show the successfully reached milestones and final results. The blocks with yellow filling show the milestones not reached by the end of the project.

### **Overview of technical approaches**

Our approach to evaluation of electron-particle collision integrals considered a significant improvement compared to the traditional methods (e. g., see (L. D. Landau, *Sov Phys. – JETP* **7**, 203-209 (1937); M. I. Kaganov, I. M. Lifshitz, L. V. Tanatarov, *Sov. Phys. - JETP*, **4**(2), 173 (1957)). The relevant electron-particle collisions are treated as interactions of the oscillating electrons with other oscillating electrons (electron-electron), polar optical phonons (electron-phonon) or defects (electron-defect) in 3D space. The oscillations are driven by electric field of a linearly polarized laser pulse. For each type of the collisions, a matrix element of the interaction is derived based on the laser-perturbed functions of Eq. (2). Accordingly, the constant 3D kinetic momentum of electrons characteristic of the traditional approaches is replaced with the 3D time-dependent canonical momentum of Eq (3). This approach implies no limitations on amplitude of laser-pulse electric field and is therefore considered as a non-perturbative one.

Following the general approach for the case of multi-cycle pulses described in Task 1, the time-dependent collision integrals are averaged over a cycle at carrier frequency, and the involved integrals are evaluated by asymptotic methods using parameter  $\alpha$  of Eq. (5). The final analytical expressions for the collision integrals  $M_{e-p}$  are represented by asymptotic series with respect to  $1/\alpha$  where the coefficients  $M_i$  incorporate analytical scaling with laser parameters:

$$M_{e-p}(t) = M_0(t) + \frac{1}{\alpha\gamma_{CB}^2} M_1(t) + \frac{1}{\alpha^2\gamma_{CB}^2} M_2(t). \quad (11)$$

Except the zero-order term, all other terms of the series contain inverse Keldysh parameter  $\gamma_{CB}$  for the conduction band. The zero-order terms of the series represent the usual non-perturbative approach under the monochromatic approximation (see V. I. Mel'nikov, *Sov. Phys. – JETP Lett.* **9** (3), 120 (1969); E. M. Epshtein, *Sov. Phys. – Solid State* **11**, 2213 (1970); A. V. Vinogradov, *Sov. Phys. – JETP*, **41** (3), 540 (1975)) where the constant amplitude of the monochromatic field is replaced with time-dependent slowly-varying pulse envelope. For example, a zero-order term of the major part of the matrix element of electron-polar-phonon interaction reads as follows:

$$M_{0e-pp}(t) = \left( \frac{eF_0 f(t/\tau_p)}{\omega_0} \right) \frac{e^2\Omega_0(\varepsilon_0 - \varepsilon_\infty)}{\pi \hbar \varepsilon_0 \varepsilon_\infty}, \quad (12)$$

where  $\Omega_0$  is polar-phonon frequency.

Three types of the collisions are considered for this Task because of their specific roles:

A) Electron-electron collisions are believed to be the fastest of all electron-particle collisions (A. Kaiser, B. Rethfeld, et al, *Phys. Rev. B* **61** (17), 11437 (2000)). Each collision of this type is usually treated as interactions of two charged particles via a screened Coulomb potential (L. D. Landau, *Sov Phys. – JETP* **7**, 203-209 (1937)). We also included spin interactions into the collision integral characteristic of the electron-electron collisions (M. I. Dyakonov, *Spin Physics in Semiconductors* (Springer- Verlag, Berlin, 2008)). Those collisions result in momentum transfer and de-phasing of the laser-driven oscillations. The traditional models suggest they negligibly contribute to light absorption since the electron-electron interactions do not accommodate the significant amount of momentum carried by absorbed laser photons. The characteristic electron-electron collision time is usually estimated by neglecting any perturbations from a laser-pulse electric field and is frequently believed to vary from 1 to 10 fs depending on specific crystals.

B) Electron collisions with polar optical phonons are the fastest among all electron-phonon interactions (A. Kaiser, B. Rethfeld, et al, *Phys. Rev. B* **61** (17), 11437 (2000); N. W. Ashcroft, N. D. Mermin, *Solid State Physics*, Saunders College Publishing, New York, 1976). Their characteristic time is usually estimated by neglecting the action of laser-pulse electric field and is believed to vary from 10 to 100 fs. They contribute to absorption of light by the conduction-band electrons since the phonons can accept the photon momentum. The phonons (characteristic energy 0.04-0.07 eV) negligibly contribute to sharing energy of absorbed photons which energy is above 0.5 eV.

C) Electron-defect collisions are negligible in crystals due to very low probability of electron interaction with defects. However, the electron-defect interactions are extremely intensive in disordered solids, e. g., glasses where the electron-defect collision time is believed to be smaller than electron-phonon collision time. This type of the collisions is included into consideration to develop a model that reasonably treats the disordered solids. The defects are assumed to be some charged centers, and the electron-defect interaction is modeled by a screened Coulomb potential.

### **Major results**

The major result of this effort included analytical relations for time-dependent collision integrals of electron-electron, electron-polar-phonon, and electron-defect collisions under the non-perturbative approximation. Those relations were utilized in the simulations of Task 3.

Among the physically meaningful results of this effort, we should mention that our estimation of collision time (that is inverse collision rate) for the electron-electron collisions vary from 10 to 100 fs depending on specific crystal. It is one order of magnitude larger than the estimations by the regular perturbative approaches. This deviation from the usual estimations is attributed to the spin interactions incorporated into our approach and correlates with the experimental data on spin relaxation in semiconductors (G. Marchetti, M. Hodgson, I. D'Amico, *J. Appl. Phys.* **116**, 163702 (2014)). Therefore, a proper accounting for the spin interactions (i. e., considering the electron-electron collisions as interactions of small magnets rather than small balls) results in a substantial increase of the electron-electron collision time. For violet-to-mid-infrared range of the optical spectrum, that collision time substantially exceeds duration of a single field cycle (and electron-oscillation cycle). This fact justifies the general low-collision-rate approach we proposed for the Task 3 when the collisions were treated as rare events that introduced small perturbations to the laser-driven oscillatory dynamics of electrons in a crystal.

Estimations of the characteristic time of the electron collisions with polar optical phonons delivered the values very close to those published by other researchers – about 100 fs.

**Task 3. Simulations of the ionization dynamics and ionization-induced optical response.**

YEAR 1:

a) Preparation and testing of the simulation code for single- and multiple-rate equations, parabolic approximation, the Keldysh formula and comparison of obtained results with published data of other authors;

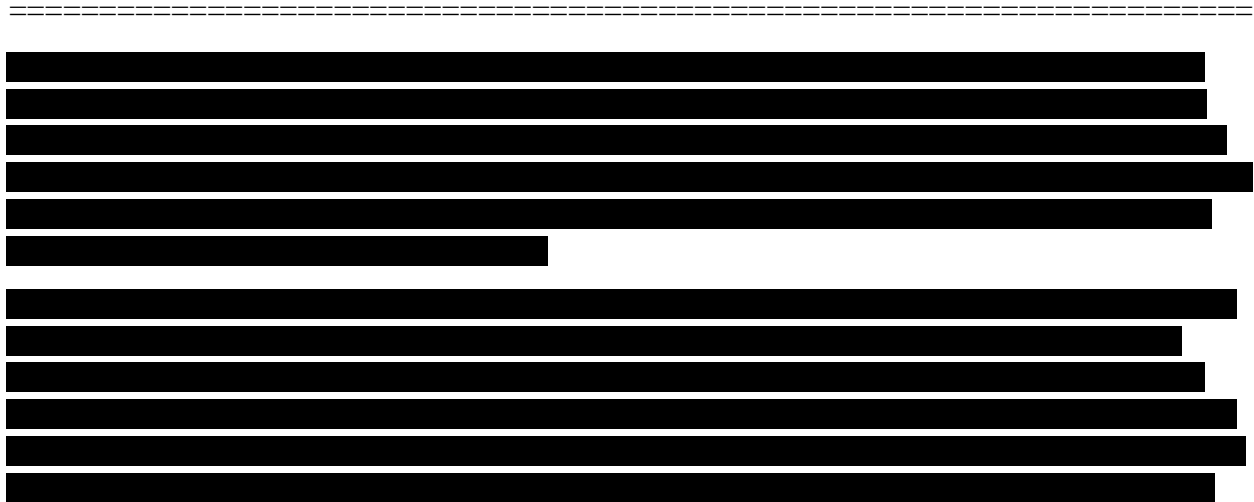
b) Simulation with the time-dependent photo-ionization rate obtained in the multi-frequency limit into the single- and multiple-rate equations with parabolic and non-parabolic bands;

YEAR 2:

c) Simulation with the time-dependent photo-ionization rate obtained for the second asymptotic limit (pulse width significantly exceeds 1 period of field oscillations) into the single- and multiple-rate equation with parabolic and non-parabolic bands; obtained results must be close to the published results for the monochromatic approximation at large pulse width;

YEAR 3:

d) Simulation of time-dependent photo-ionization rate obtained for the first asymptotic limit (pulse width close to 1 period of field oscillations) into the single- and multiple-rate equation with parabolic and non-parabolic bands.



This Task incorporates the deliverables from Task 1 (the photoionization rate) and Task 2 (the rates of electron-particle collisions). In contrary to the two other tasks, this Task does not include analytical calculations and is focused on numerical solving some equations for electron density in the conduction band. It delivers ultrafast material response to action of ultrashort laser pulses that can serve as input to model transient optical properties for propagation equations in a simulation of ultrafast nonlinear propagation. The specific outputs of this Task include:

- I) Instant and total density of conduction-band electrons;
- II) Instant and total amount of laser-pulse energy absorbed via nonlinear mechanisms, e. g., multiphoton absorption attributed to the inter-band electron transitions;
- III) Instant and total amount of laser-pulse energy absorbed by the conduction electrons via the intra-band excitation within the conduction band that scales linearly with laser intensity;
- IV) Rates of the linear and nonlinear (with respect to laser intensity) absorption;
- V) Transient linear and nonlinear optical response during a direct action of a laser pulse.

## Overview of technical approaches

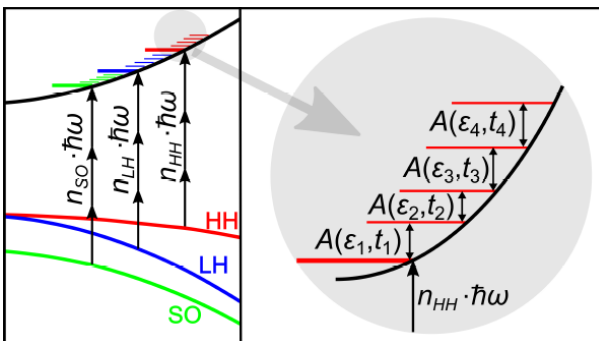
The objective of this task was to simulate electron dynamics using the rates of electron excitation and collisions from Tasks 1 and 2. A regular rate equation for conduction-band electron density:

$$\left( \frac{d N_{CB}}{d t} \right) = W_{PI} + W_{IMP} - W_R, \quad (13)$$

is a minimum and the most popular approach to do that (see, e. g., S.S.Mao, et al, *Appl. Phys. A* **79**, 1695 (2004); P. Balling and J. Schou, *Rep. Prog. Phys.* **76**, 036502 (2013)). In Eq. (13), the right-hand part contains the rates of photoionization  $W_{PI}$ , impact ionization  $W_{IMP}$ , and electron recombination  $W_R$ . Since Eq. (13) is frequently utilized to simulate ultrafast optical response of solids (L. Sudrie, A. Couairon, et al, *Phys. Rev. Lett.* **89**, 186601 (2002); J. R. Gulley, W. M. Dennis, *Phys Rev. A* **81**, 033818 (2010); A. Couairon, A. Mysyrowicz, *Phys. Reports* **441**, 47-189 (2007).), it was employed in the simulations for this Task. Alternative approaches consider numerical solving a Fokker-Plank equation (T. Apostolova, Y. Hahn, *J. Appl. Phys.* **88** (2), 1024-1034 (2000)). The most rigorous approach is to solve the Boltzmann equation (A. Kaiser, B. Rethfeld, et al, *Phys. Rev. B* **61** (17), 11437-11450 (2000)). Simulations with those approaches involve the kinetic coefficients (i. e., the rates of the electron excitation) and are considered as the next step to continue this effort in a separate project.

The simplest rate equation (13) is improved according to the results obtained from Tasks 1 and 2. First, electron transitions from several valence bands are taken into account. In the first approximation, electron excitation between the valence bands is prohibited (N. W. Ashcroft, N. D. Mermin, *Solid State Physics*, Saunders College Publishing, New York, 1976). Therefore, the valence bands independently contribute to the buildup of the conduction-band electron density. Correspondingly, three photoionization rates are introduced in the right-hand part of Eq. (13) for the electron transitions from heavy-hole (HH), light-hole (LH), and split-off (SO) valence bands (Fig. 4). Second, the laser-generated free electrons arrive in the conduction band at different values of energy (Fig. 4). Therefore, the coefficients of the right-hand part of Eq. (13) introduce dependence on instant energy of conduction-band electrons. Third, depopulation of the valence bands must be taken into account since it reduces the rate of the electron excitation. Finally, the results from Task 2 suggest that the rate of electron-phonon collisions is low enough to suppress the impact ionization. This suggestion is also confirmed by the negative results of experimental attempts to detect the impact ionization in crystals (A. Mouskeftaras, S. Guizard, et al, *Appl. Phys. A* **110**, 709 (2013)). Therefore, the revised rate equation of Eq. (13) reads as follows:

$$\frac{d N_{CB}}{d t} = f_{HH}(t) \cdot W_{HH} [I(t)] + f_{LH}(t) \cdot W_{LH} [I(t)] + f_{SO}(t) \cdot W_{SO} [I(t)], \quad (14)$$



**Fig. 4.** A sketch of intra-band electron dynamics. Three valence bands are shown to be specific – heavy-hole (HH - red), light-hole (LH - blue), and split-off (SO – green).  $n_{SO}$ ,  $n_{LH}$ , and  $n_{HH}$  are the minimum number of laser photons at carrier frequency required to bridge the energy gap between the conduction band and the valence bands SO, LH, and HH respectively.  $A(\epsilon, t)$  is the rate of energy absorption in the conduction band.

where  $f_{HH}(t)$ ,  $f_{LH}(t)$ , and  $f_{SO}(t)$ , are the population coefficients determined from populations of the valence bands and the conduction band as follows:

$$f_{HH}(t) = \frac{N_{HH} - N_{CB}^{HH}(t)}{N_{HH}}; \quad f_{LH}(t) = \frac{N_{LH} - N_{CB}^{LH}(t)}{N_{LH}}; \quad f_{SO}(t) = \frac{N_{SO} - N_{CB}^{SO}(t)}{N_{SO}}. \quad (15)$$

and  $W_{HH}$ ,  $W_{LH}$ , and  $W_{SO}$  are the photoionization rates for the transitions from HH, LH, and SO valence bands to the conduction band correspondingly. The total conduction band population consists of three contributions:  $N_{CB}(t) = N_{CB}^{HH}(t) + N_{CB}^{LH}(t) + N_{CB}^{SO}(t)$ .

With clear understanding of invalidity of the usual Drude model to properly describe absorption by free electrons and dynamics in the conduction band (see, e. g., A. V. Vinogradov, *Sov. Phys. – JETP* **43** (3), 521 (1976)), it was proposed to replace the Drude model with the Vinogradov equation (A. V. Vinogradov, *Sov. Phys. – JETP* **41** (3), 540 (1975)). According to that equation, the average rate of energy absorption by a single conduction electron receives contributions from collisions of polar optical (the second right-hand term) and acoustic (the first right-hand term) phonons with the oscillating electrons:

$$\begin{aligned} \left( \frac{d\varepsilon}{dt} \right)_{CB} &= \left( \frac{eF(t)}{\omega_0} \right)^3 \frac{2G^2 k_B T_{phonon}}{\pi^2 \hbar^4 \rho u^2} \Psi_1 \left( \frac{\omega_0 \sqrt{2m\varepsilon(t)}}{eF(t)} \right) \\ &+ \left( \frac{eF(t)}{\omega_0} \right) \frac{e^2 \Omega_0 (\varepsilon_0 - \varepsilon_\infty)}{\pi \hbar \varepsilon_0 \varepsilon_\infty} \coth \left( \frac{\hbar \Omega_0}{2k_B T_{phonon}} \right) \Psi_2 \left( \frac{\omega_0 \sqrt{2m\varepsilon(t)}}{eF(t)} \right) \end{aligned} \quad (16)$$

Here  $\varepsilon_0$  and  $\varepsilon_\infty$  are static and high-frequency dielectric constants;  $F(t)$  is electric field of the slowly-varying pulse envelope;  $\varepsilon(t)$  is time-dependent energy of the electrons;  $T_{phonon}$  is phonon temperature,  $\rho$  is density,  $\Omega_0$  is polar-phonon frequency, and  $G$  is deformation potential. The two  $\Psi$  functions of the right-hand part of the equation are defined in the paper of Vinogradov. Eq. (16) is derived from the Boltzmann equation under the classical approximations that underlie the classical approach of the Drude formula. However, Eq. (16) is derive by assuming no limitations on amplitude of laser-pulse electric field and by assuming that the rate of electron-particle collisions is significantly smaller than carrier frequency of a laser pulse. Also, Eq. (16) considers parabolic energy-momentum relations both for the conduction electrons and colliding phonons. An equation similar to Eq. (16) was derived for a cosine conduction band and was utilized in the line of simulations focused on cosine-band approximation with proper rates from Tasks 1 and 2.

The absorption rate of Eq. (16) is introduced into the equation for the total energy absorption:

$$\begin{aligned} \frac{dE}{dt} &= f_{HH}(t) \cdot W_{HH}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{HH}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega + f_{LH}(t) \cdot W_{LH}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{LH}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega \\ &+ f_{SO}(t) \cdot W_{SO}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{SO}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega + f_{PE1}(t) \cdot W_{PE1}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{PE1}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega + \left( \frac{d\varepsilon}{dt} \right)_{CB1}, \end{aligned} \quad (17)$$

where the first three terms describe energy absorption due to the photoionization of electrons residing in the heavy hole (HH), light-hole (LH), and split-off (SO) valence bands respectively.

The overall approach considered numerical solving the coupled equations (14) – (16) (O. Sergaeva, V. Gruzdev, *JOSA B* **35** (11), 2895 (2018)). In parallel, Eq. (17) was solved to deliver both total and instant values of linear (the last term of Eq. (17)) and nonlinear absorption. Linear and nonlinear imaginary parts of transient optical response were obtained from a solution to Eq.

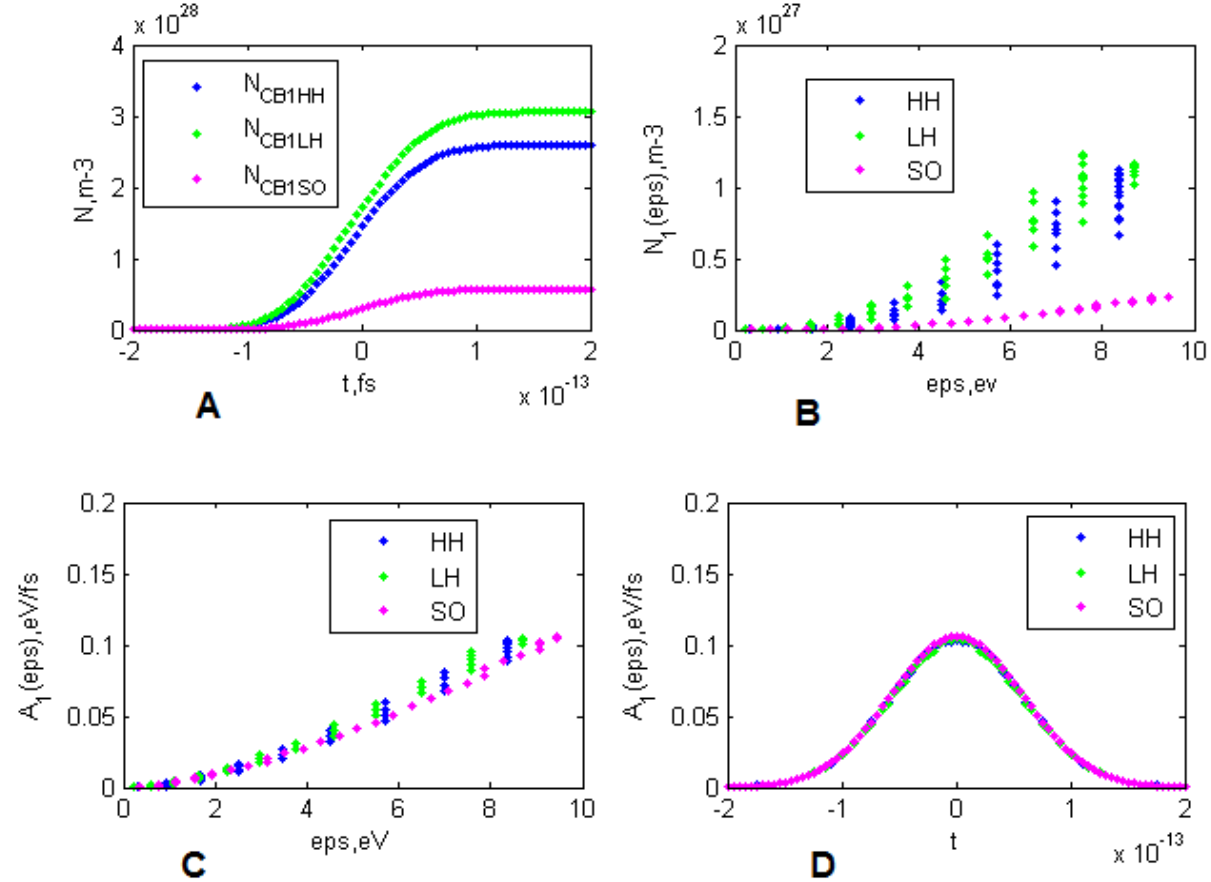
(17). Linear and nonlinear real parts of the transient optical response were obtained via Kramers-Kronig relations under the assumption that the spectrum of absorption was limited by the spectrum of a laser-pulse. For comparison, similar simulations are done for the usual Drude model frequently employed in the rate-equation simulations (S.S.Mao, et al, *Appl. Phys. A* **79**, 1695 (2004); P. Balling and J. Schou, *Rep. Prog. Phys.* **76**, 036502 (2013)). It delivers the following equation for the rate of energy absorption by the conduction-band electrons:

$$\left(\frac{d\varepsilon}{dt}\right)_{DR} = \frac{e^2 F_0(t)^2}{2m_{CB}\omega^2} \frac{\omega^2}{\omega^2 + \nu_{DR}^2} \nu_{DR}, \quad (18)$$

where  $\nu_{DR}$  is an electron-particle collision rate. For the simulations, the Drude collision rate was assumed to be  $10^{15}$  1/s following the results obtained by fitting some experimental data with the Drude model (S.S.Mao, et al, *Appl. Phys. A* **79**, 1695 (2004)). Details of the simulations have been published (see O. Sergaeva, V. Gruzdev, *JOSA B* **35** (11), 2895 (2018)).

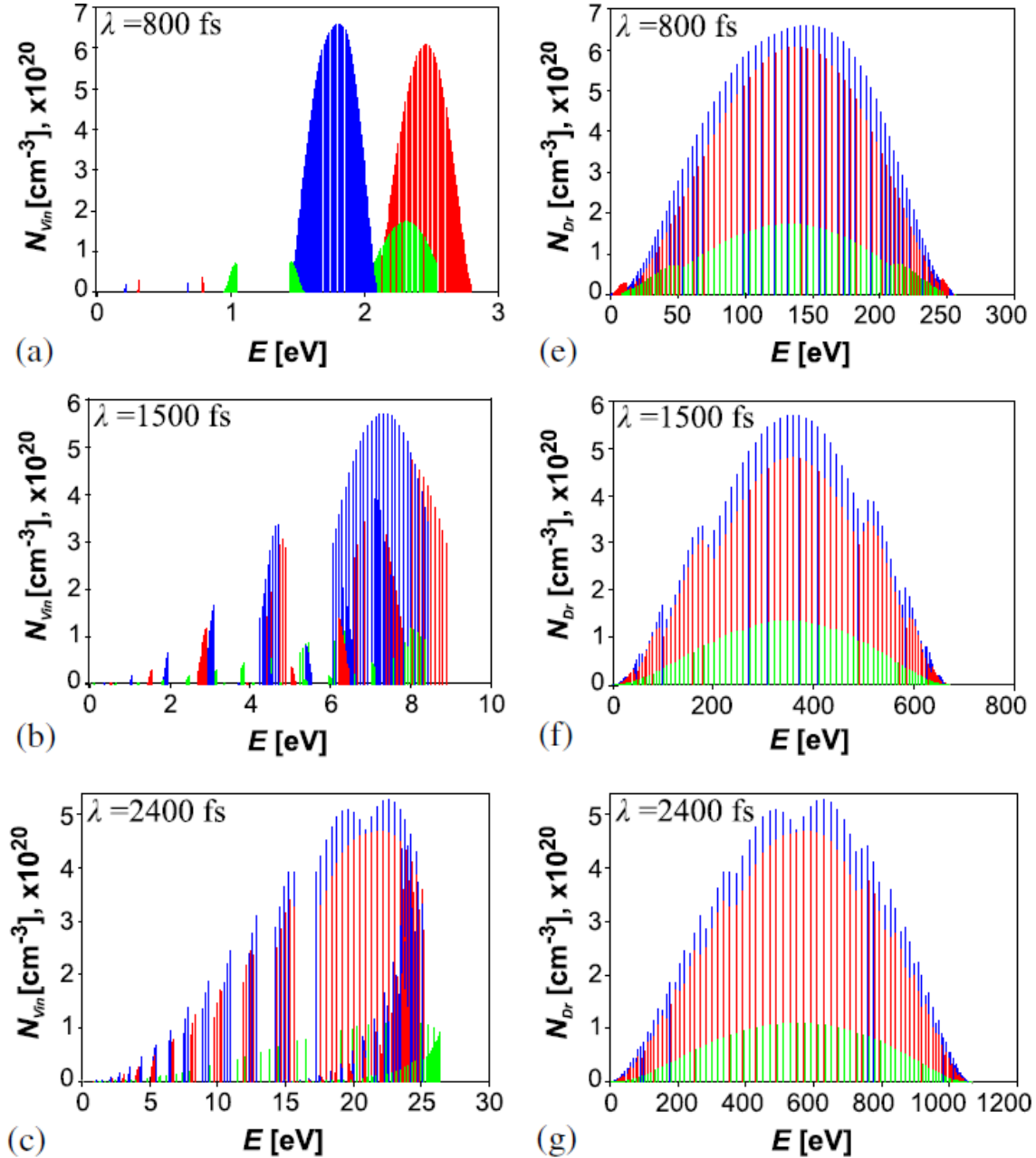
### Major results

In contrary to the usual approaches based on a single rate (S.S.Mao, et al, *Appl. Phys. A* **79**, 1695 (2004)) or multiple rate (B. Rethfeld, *Phys. Rev. Lett.* **92**, 187401 (2004); B. Rethfeld, *Phys. Rev. B* **73**, 035101 (2006)) equations, our model delivers a detailed information about electron dynamics driven by ultrashort laser pulses including electron energy distribution (Fi. 5).



**Fig. 5.** Dependence of conduction-electron density on time (A); energy distribution of conduction electrons (B); energy dependence of conduction-electron absorption rate (C); and time evolution of conduction-electron absorption (D) simulated for ZnSe interacting with 100-fs laser pulse at wavelength 2000 nm and peak irradiance 10 TW/cm<sup>2</sup>. HH, LH, and SO mark the values obtained for heavy-hol, light-hole, and split-off valence bands respectively.

This simple model predicts non-equilibrium energy distribution of conduction-band electrons (Figs. 5 B and 6) that is qualitatively similar to early stages of electron dynamics delivered by the Boltzmann kinetic equation (A. Kaiser, B. Rethfeld, et al, *Phys. Rev. B* **61** (17), 11437 (2000)). The specific structure of the energy distribution suggested to pay more attention to simulation of the energy distribution (see O. Sergaeva, V. Gruzdev, *JOSA B* **35** (11), 2895 (2018)) and comparing it to the one of the high-collision-rate Drude model (fig. 6). Several non-trivial results were obtained under the approximation of parabolic conduction band.



**Fig. 6.** Electron distributions in ZnSe conduction band at the tail ( $t = 200$  fs) of 100-fs laser pulse at  $10 \text{ TW/cm}^2$  peak irradiance at several values of wavelength: (a) and (e) - 800 nm; (b) and (f) - 1500 nm; (c) and (g) - 2400 nm; Panels (a) through (c) are for the Vinogradov equation. Panels (e) through (g) are for the Drude equation. Separate contributions of each valence band are depicted by red for HH, blue – for LH, and green – for SO. The bottom of the conduction band is assumed to be at zero level.

The energy distributions simulated with the Vinogradov equation cannot be characterized by electron temperature and demonstrate two specific features: reduced high-energy tail of the distribution and splitting of the entire conduction-band population into several separated sub-populations. The high-energy cut-off of the distributions is explained by a competition between the rate of intra-band promotion of the electrons according to Eq. (16) and the rate of effective-band-gap modification by the ponderomotive energy of the laser-driven oscillations incorporated into the photoionization rates of Eq. (14). The modification of effective band gaps results in a step-wise time variation of the number of simultaneously absorbed photons to bridge an instant effective band gap. For the Vinogradov model, the effective band gap is modified faster than the conduction electrons are promoted to higher energy by the collisions. The generation of new sub-populations by the step-wise modification of the number of simultaneously absorbed photons produces the cut-off because it is much faster than the promotion of existing sub-populations to higher energy that produces a gradient reduction of the high-energy tail.

Formation of the separated sub-populations reasonably results from the step-wise modification of the arrival energy because of the step-wise changes of the number of simultaneously absorbed photons. That process is enhanced by simultaneous contributions from several valence bands and dominates over spreading of the sub-populations by the low-rate electron-phonon collisions (see O. Sergaeva, V. Gruzdev, *JOSA B* **35** (11), 2895 (2018)). Qualitatively, formation of the sub-populations agrees with generation of peaks of non-equilibrium electron distributions simulated by a full Boltzmann equation (A. Kaiser, B. Rethfeld, et al, *Phys. Rev. B* **61** (17), 11437 (2000)). The peaks were utilized to justify introduction of the multiple-rate equations for the conduction-band electron density (B. Rethfeld, *Phys. Rev. Lett.* **92**, 187401 (2004)). However, the energy levels of the multiple-rate equations are artificially picked up and are equidistant with a gap of one photon energy between them. Our results show this is not the case, e. g., the sub-populations are not equidistant. The model of multiple-rate equation is based on a questionable physical concept.

The Drude model delivers highly equilibrium distributions of electrons because the high-rate collisions rapidly equilibrate the conduction-band population. Conceptually, it fits the model of the impact ionization very well. However, the impact ionization was not detected in experiments on crystals. Also, a direct consequence of the high-rate collisions is generation of a keV-energy tail of the distributions (fig. 6). This result suggests emission of electrons with keV energy from the dielectrics excited by ultrashort pulses at intensity below damage threshold. Such energies are not observed in experiments (A. N. Belsky, H. Bachau, et al, *Appl. Phys. B* **78**, 989-994 (2004)).

Among significant physical results predicted by our simulations are formation of the isolated sub-populations in the conduction band and inversion of conduction-band population (Fig. 6). Since the sub-populations can survive within several collision times, i. e., for the time interval from few hundreds of fs to few picoseconds, those two effects suggest possible amplification of weak probe pulses by the conduction-band electrons excited by an ultrashort (150 fs or shorter) pump pulse. It is remarkable that amplification of a probe pulses was recently reported in dielectrics (Th. Winkler, et al, *Nature Physics* **14**, 74 (2018)) and received no adequate interpretation. Moreover, our simulations for a cosine band predict a specific amplification of probe pulses by the laser-driven electron oscillations. Qualitatively similar effects were observed in non-collision plasmas some 50 years ago, but they have never been reported for solids.

Results obtained from this Task have been reported in our recent publication (O. Sergaeva, V. Gruzdev, *JOSA B* **35** (11), 2895 (2018)) and will appear in a few more papers either submitted or being prepared for submission to peer-reviewed journals.

## CHALLENGES AND BOTTELNECKS

[REDACTED]

4) Among the challenges, I would mention a specific treatment of our peer-reviewed journal submissions by reviewers. It took 16 months to publish our Phys. Rev. B paper with 5 rounds of review and 6 reviewers. A majority of responses we received were not relevant to scientific and technical merits of our work, for example: another model of the photoionization was not needed; an analytical model was not needed because of a broad class of numerical approaches available to simulate ionization dynamics. Also, we received many comments of the reviewers either addressing not relevant physical effects or wrongly interpreting some fundamental solid-state physics. I understand that we are delivering new physics that contradicts the common “religions” in the field of ultrafast laser-solid interactions. For example, we justified invalidity of the combination of the Keldysh formula with the high-collision-rate Drude model, but that combination has been reported in hundreds of previous publications. However, this situation signals about substantial degradation of qualification of reviewers in the specific field of science and makes me aware of a slow and painful review process for all our manuscripts.

## CONCLUSIONS

Overall, the research effort supported by this award was extremely successful in spite of the several milestones not reached by the end of year 3. Our efforts delivered groundbreaking results and some of them were not expected at the beginning of the project. In fact, results of this project have totally changed the landscape of theoretical models for high-intensity interaction of few-cycle (from 3 to 30 cycles) laser pulses with dielectric solids.

First, we have developed a theory of the photoionization for two the most meaningful cases: a single-cycle laser pulse and a multi-cycle pulse containing from 3 to 30 cycles. We have demonstrated that pulses longer than 25-30 cycles can be properly treated under the regular monochromatic approximation, e. g., by the Keldysh formula for the photoionization rate. Moreover, we have delivered an analytical criterion (that contains a combination of laser and material parameters) to judge if the monochromatic approximation is acceptable for specific values of laser and material parameters. Our relations deliver scaling of the photoionization rate and relevant effects with 6 parameters including central wavelength, pulse width, peak intensity, carrier-envelope phase, band gap, and effective electron and hole masses. This is very valuable supplement for the numerical simulations of the ultrafast laser-induced electron excitation.

Second, we have derived non-perturbative time-dependent relations for electron-particle collision rates in both parabolic and cosine conduction bands for the case of multi-cycle pulses. In contrary to the previously published expressions, our relations do not put limitations on amplitude of electric field, do not assume single frequency in a laser-pulse spectrum, and consider arbitrary pulse envelope.

Third, we have fixed several fundamental contradictions of the most popular models of laser-driven electron dynamics based on single or multiple rate equations. That approach is widely used in simulations of ultrafast material response and absorption for nonlinear propagation, laser-induced damage, and ultrafast laser ablation. The traditional approaches combine the high-collision-rate Drude model (with electron-particle collision rates of  $10^{15}$  1/s and higher) of electron heating in the conduction band with zero-collision Keldysh formula for the rate of inter-band electron transitions. Either the Keldysh formula or the Drude model should be removed from that combination to fix that contradiction. Also, the Drude model is based on the assumption that electric-field amplitude is low enough to use a usual perturbation theory. The Keldysh formula employed in those contradictive models did not imply limitations on value of the electric field. Finally, the Keldysh formula was employed in its original version that included electron transitions from a single valence band only. To fix those contradictions, we have:

- incorporated contributions from several valence bands to the photoionization transitions;
- replaced the high-collision-rate Drude model with low-collision-rate Vinogradov equation that was derived under the non-perturbative approach;
- incorporated dependence of the absorption by free electron on their energy in the conduction band;
- replaced the collision time of the Drude model (that was essentially utilized as a fitting parameter) with microscopic relations for matrix elements of electron-phonon interactions.

We also note that the photoionization and electron-particle collision rates derived under the support of this award can be utilized as kinetic coefficients in any approach to simulation of the laser-driven electron dynamics in the conduction band, e. g., by a single rate equation, the

Fokker-Plank equation, or the Boltzmann equation. They can also be incorporated into more general quantum or classical kinetic equations. Therefore, we have developed a universal basis for a broad range of simulations of the laser-driven electron dynamics.

Among the groundbreaking “side” results suddenly produced by this effort I would mention the following:

- prediction of generation of cycle-averaged bias-free photocurrent in dielectrics and wide-band-gap semiconductors. Since it is very sensitive to carrier-envelope phase, the photocurrent can be used to measure CEP by a compact desk-top device. This effect can deliver novel approaches to control and shape ultrashort laser pulses and also can contribute to ultrafast nonlinear optical response.
- prediction of amplification of weak probe pulses by the conduction-band electrons excited by high-intensity pump pulses with sub-150-fs duration.
- deformation of cosine conduction band that produces modifications of original effective mass by electric field of ultrashort laser pulses. Since the effective mass affects all dynamic properties and responses of electrons in the solids, this effect opens a novel approach to laser control over electronic processes in dielectrics and wide-band-gap semiconductors. Also, this effect can contribute to the ultrafast nonlinear response of those solids that affect nonlinear propagation.
- formation of sub-populations of the conduction-band electrons produces electron distributions qualitatively similar to those observed in experiments on above-threshold ionization of gases. This fundamental fact can substantially contribute to the recently discovered fundamental similarity between ultrafast strong-field effects in atomic and solid-state quantum systems.

Obtained results have been delivered to a broad research community via 14 presentations at international conferences, 4 seminar talks, 5 conference papers, and two manuscripts published in *Physical Review B* and *Journal of the Optical Society of America B*. Currently, we have several more manuscripts either already submitted to peer-review journals or being prepared for submission to the journals.

Our results have produced a very strong response by US and international researchers. It is remarkable that our paper published in *Phys. Rev. B* in September 2018 has received 3 citations within the first month after the publication. Success of the conference presentations and the feedback of a target research community received at the conferences have resulted in several collaborations with US research groups. Our results have stimulated international collaborations with researchers from France and Germany. Those domestic and international research contacts have very high potential for spinning out joint research projects, grant applications, and publications.

However, publishing our novel results is very difficult because they clearly demonstrate that a majority of published simulations of the ultrafast laser-crystal interactions are wrong and utilize the contradictive models beyond the ranges of their validity. Many researchers (who are reviewers of our papers) are not ready to recognize that fact.

The only issue that substantially affected our research under this award is the mistake with hiring an improper postdoc for the first year of this project. However, the measures undertaken to mitigate that issue were quite successful and minimized the impact of that issue.